

Docket No.: 4569D (DIV)  
(PATENT)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

|                                     |                               |
|-------------------------------------|-------------------------------|
| In re Patent Application of:        | :                             |
| Hoke et al.                         | :                             |
|                                     | :                             |
| Application No.: 10/002,272         | : Group Art Unit: 1764        |
|                                     | : Examiner: Jennifer A. Leung |
| Filed: November 15, 2001            | :                             |
|                                     | :                             |
| For: METHOD AND DEVICE FOR CLEANING | :                             |
| THE ATMOSPHERE                      | :                             |
|                                     | :                             |
|                                     | :                             |
|                                     | :                             |

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APPEAL BRIEF

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### **Real Party in Interest**

The real party in interest in this proceeding is BASF Catalysts LLC, the predecessor in interest to the Assignee of record, Engelhard Corporation.

### **Related Appeals and Interferences**

Neither Appellant nor its agents are aware of any prior or pending appeals, judicial proceedings or interferences which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

### **Status of Claims**

Claims 1-36 and 54 have been cancelled, and claims 37-47 have been withdrawn. Claims 48-53 and 55-58 stand finally rejected under 35 U.S.C. § 103(a) and 35 U.S.C. § 112 and are appealed. A copy of the claims on appeal are in the Claim Appendix of this Brief.

### **Status of Amendments**

A Response to Notice of Noncompliant Amendment submitted on January 12, 2007 was entered by the Examiner, as stated in an Advisory Action mailed on February 1, 2007.

### **Summary of Claimed Subject Matter**

Claim 48 is directed to a device for treating the atmosphere to catalytically convert atmospheric pollutants to less harmful materials comprising an outer surface of a motor vehicle component 100 which is exposed to a flow of ambient air, the outer surface

being coated with a catalyst composition 102 that is protected with a porous overcoat of carbon 104. (Page 4, line 19 through page 5, line 25; Fig. 1A.)<sup>1</sup>

Claim 49 depends from claim 48, and further requires one hydrophobic protective material 104 overcoating the overcoat of carbon, the protective material capable of substantially preventing liquid water and/or water vapor from reaching the catalytic composition or overcoat of carbon. (Page 6, lines 21-24; Fig. 1A.)

Claim 50 depends from claim 48, and further requires at least one layer of the hydrophobic material overcoating the catalyst composition or overcoat of carbon. (Page 6, lines 6-7.) Claim 51 depends from claim 48 and further requires that the catalyst composition is selected from base metals, precious metals, salts and oxides thereof and combinations thereof. (Page 13, line 21 through page 14, line 7.) Claim 52 depends from claim 48 and further requires that the catalyst comprises manganese dioxide. (Page 13, lines 23-24.) Claim 53, depending from claim 48, requires that the overcoat of carbon is selected from activated granular carbon, carbon black, permanganate on carbon, and mixtures thereof. (Page 12, lines 24-26.) Claim 55 depends from claim 49 and requires that the hydrophobic material is selected from fluoropolymers and silicone polymers. (Page 13, lines 4-19.)

Claim 56 depends from claim 48 and recites that the outer surface of the motor vehicle component is selected from the outer surface of radiators, air-conditioner condensers, charge air coolers, transmission coolers and inserted devices. (Page 5, lines 20-24.) Claim 57 depends from claim 48 and specifies that the catalyst composition is selected from manganese dioxide, platinum, palladium and mixtures thereof. (Page 6,

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<sup>1</sup> Page and paragraph designations refer to those found in the originally filed specification.

lines 25-28.) Claim 58 depends from claim 48 and specifies that the catalyst composition is supported on a material selected from ceria, alumina, titania, silica, zirconia and mixtures thereof. (Page 14, lines 9-13.)

### **Grounds of Rejection to be Reviewed on Appeal**

1. Whether claims 48-53 and 55-58 are unpatentable under 35 U.S.C. § 112, second paragraph, as being indefinite.

2. Whether claims 48, 51-53 and 56-58 are unpatentable under 35 U.S.C. § 103(a) over WO 98/022235 in view of JP 52-122290.

3. Whether claims 49, 50 and 55 are unpatentable under 35 U.S.C. § 103(a) over WO 98/022235 in view of JP 52-122290 further in view of Okamoto et al JP 55-023039.

Although *not* part of the scope of this appeal, Appellants respectfully point out that claims 48-53 and 55-58 in this application stand rejected under the judicially created doctrine of obviousness-type double patenting as allegedly unpatentable over claims 6,190,627 in view of JP 52-122290. As stated in an amendment submitted March 14, 2005, Appellants are prepared to file a terminal disclaimer upon allowance of at least one pending claim.

### **Argument I**

Claims 48-53 and 55-58 in this application stand finally rejected under 35 U.S.C. § 112, second paragraph, as allegedly being indefinite.

#### **Claim 48**

In the office action mailed August 24, 2006 ("Office Action"), the Examiner rejected the claims, stating "[i]n claim 48, line 3 it is unclear as to how the ambient air is related to the atmosphere set forth in line 1." (Office Action, Page 2, paragraph 3.)

The Manual of Patent Examining Procedure requires that “the words of the claim must be given their plain meaning unless applicant has provided a clear definition in the specification.” (MPEP § 2111.01 (citations omitted.)) Accordingly, it is fundamental under 35 U.S.C. § 112, second paragraph that Appellants can be their own lexicographers and that they “can define in the claims what they regard as their invention essentially in whatever terms they choose so long as any special meaning assigned to a term is clearly set forth in the specification.” (MPEP § 2173.01.) The MPEP cautions that examiners “should allow claims which define the patentable subject matter with a reasonable degree of particularity and distinctness” and that “[s]ome latitude in the manner of expression and the aptness of terms should be permitted even though the claim language is not as precise as the examiner might desire.” (MPEP § 2173.02.)

In the amendment submitted on March 14, 2005, Appellants pointed out that the specification clearly establishes the relationship between the terms “atmosphere” and “ambient air” in the application. In the specification, “ambient air” is defined as “the portion of the atmosphere that drawn or forced toward the outer surface of the coated substrate” (see page 5, lines 12-13 of the specification). The specification defines “atmosphere” as “mass of air surrounding the earth” (see page 5, lines 10-11 of the specification).

Appellants respectfully submit that in view of the definitions in the specification, the relationship between “atmosphere” and “ambient air” is clear. That is, ambient air is a portion of the atmosphere that comes towards the coated substrate. Since the language of the claim is such that a person of ordinary skill in the art could interpret the metes and bounds of the claim so as to understand how to avoid infringement, the rejection of the

claim under 35 U.S.C. § 112, second paragraph, is inappropriate. See MPEP § 2173.02. Claim 48, therefore, is not indefinite.

### **Claims 49-53 and 55-58**

For at least the reasons provided above with respect to claim 48, claims 49-53 and 55-58 are not indefinite.

### **Argument II**

Claims 48, 51-53 and 56-58 stand finally rejected as allegedly unpatentable under 35 U.S.C. § 103(a) over WO 98/02235 in view of JP-52-122290.<sup>2</sup>

### **Claim 48**

The Office Action cites WO 98/02235 as disclosing a motor vehicle component, which is exposed to a flow of ambient air, the radiator being coated with a catalyst material selected from a base metal, a precious metal or manganese oxide and a protective layer of polymers. (Office Action, Page 3, paragraph 3.) The Office Action admits that WO 98/02235 is silent as to the protective coating comprising a porous overcoat of carbon. (Office Action, Page 3, paragraph 4.)

The Office Action relies upon JP52-122290 for the teaching of the protection of a catalyst composition with porous overcoat of carbon. (Office Action, Page 3, paragraph 4.) The Office Action concludes that it would have been obvious to provide a porous overcoat of carbon material on the surface of the catalyst material as taught by JP 52-122290 as a substitute for the porous protective layer of polymers in the apparatus of WO 98/02235 because the porous overcoat of carbon is allegedly capable of protecting the

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<sup>2</sup> The Office Action cited JP 52-122290, but only the Abstract for this document was provided, contrary to the requirements of MPEP §706.02. In order to create a complete record, however, Appellants are submitting a copy of a translation of JP 52-122290 with this Appeal Brief.

catalyst composition form poisoning and degradation from a variety of contaminants, and that the substitution of one known protective coating for another known protective coating merely involves routine skill in the art. (Office Action, Pages 3-4, paragraph 4.)

To establish a *prima facie* case of obviousness, three basic criteria must be met. There must be some suggestion or motivation from the references themselves or the knowledge generally available to one of ordinary skill in the art to modify the reference or to combine reference teachings. Second, there must be a reasonable expectation of success. The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be in the prior art, and not in the Appellants' disclosure. Third, prior art references must teach or suggest all the claim limitations (MPEP § 2142.)

In the instant case, Appellants respectfully submit that the Office Action fails to establish *prima facie* obviousness. Appellants recognize that WO 98/02235 teaches a catalyzed surface of a motor vehicle component to convert atmospheric pollutants such as CO to less harmful materials. However, WO 98/02235 teaches the use of a resistant, repellent overcoat to protect the underlying catalyst from roadway contaminants such as chemicals, salts, dirt and other contaminants which may adversely affect the catalyst composition. (WO 98/02235, Page 15, lines 25-33.)

JP 52-122290 teaches the use of an adsorptive overcoat material to protect an exhaust gas catalyst used to purify a waste gas stream from automobiles, combustion furnaces, oil burners, incinerators and chemical plants to protect the catalyst from waste gas stream contaminants such as S, P, Pb, Ca, and Ni. The Office Action states that it would have been obvious to substitute the polymeric, resistive porous protective layer in WO 98/02235 with the porous carbon overcoat of JP 52-122290 on the surface of the

catalyst material to prevent catalyst poisoning and degradation from a variety of contaminants.

Appellants respectfully submit that the skilled artisan reading WO 98/02235 together with JP 52-122290 would not be motivated to substitute the adsorptive coatings disclosed in JP 52-122290 for the resistive or repellent coating disclosed in WO 98/02235 to repel chemicals, salts, dirt and other contaminants. First, WO 98/02235 pertains to the field of treating the atmosphere by contacting the catalyst contained on the surface of an automobile with ambient air containing pollutants to convert the pollutants into harmless compounds. There is no teaching or suggestion in WO 98/02235 to provide an adsorptive coating over the catalyst to prevent catalyst poisoning. To do so would render the device taught in the reference unsuitable for its intended purpose, which is to provide an article with a coating that repels contaminants such as chemicals, salts, dirt and other contaminants from the catalyst surface. If the proposed modification of a reference renders the reference unsuitable for its intended purpose, there can be no suggestion to make the proposed modification. See MPEP § 2143.01 Subheading V.

Furthermore, JP 52-12290 pertains to catalysts for the treatment of waste gases, from automobiles, combustion furnaces, oil burners, incinerators and chemical plants, not catalysts for the treatment of ambient air. The various coatings in JP 52-12290 are described as being useful for protecting the catalyst from S, Pb, Ca, and Ni in a waste gas stream. Appellants submit that it cannot be assumed that a material suitable for treating a waste gas stream would and for adsorbing S, Pb, Ca and Ni would be suitable as a protective overcoat for a catalyst used in the treatment of ambient air to repel chemicals, salts, dirt and other contaminants because the two types of catalysts encounter different

temperatures, pollutant mix/concentration and other use conditions and the protective coatings have completely opposite functions. There is no teaching or suggestion in JP 52-12290 that a porous carbon coating would be suitable for protecting a catalyst the types of contaminants encountered in WO 98/02235, namely, chemicals, salts, dirt and other contaminants. It appears that the Office Action has relied on improper hindsight in assuming that the carbon coating used in a waste gas stream for protecting the catalyst from different types of contaminants would be successful in protecting the catalyst in the instant invention. There is no teaching in JP 52-12290 that a carbon coating would be useful to coat a catalyst for treating the atmosphere, and thus there is no reasonable expectation of success that the coating in JP 52-12290 would protect the catalyst in WO 98/02235 from chemicals, salts, dirt and other contaminants. Accordingly, due to the failure to provide a proper motivation to combine, *prima facie* obviousness has not been established, and the combination of WO 98/02235 and JP 52-12290 cannot render claim 48 obvious.

**Claims 49, 51-52, and 56-58**

For at least the reasons provided above with respect to claim 48, claims 49, 51-52, and 56-58 are not obvious over the cited art.

**Claim 53**

Regarding claim 53, claim 53 recites specific types of carbon, namely, activated granular carbon, carbon black, permanganate on carbon and mixtures thereof. The Office Action fails to specifically address the limitations of claim 53. There is no teaching or suggestion in WO 98/02235 or JP 52-122290 of the particular types of carbons recited in claim 53. Because the Office Action fails to recite prior art containing all of the claimed

elements of claim 53, *prima facie* obviousness has not been established. The rejection of claim 53, therefore, should be reversed.

### **Argument III**

Claims 49, 50 and 55 stand finally rejected as allegedly being unpatentable under 35 U.S.C. § 103(a) over WO 98/02235 in view of JP 52-122290 as applied to claim 48 and further in view of JP 55-023039.

### **Claims 49 and 50**

The Office Action admits that the collective teaching of WO 98/02235 and JP 52-12290 does not teach or suggest at least one layer of a hydrophobic protective material overcoating the overcoat of carbon, wherein the hydrophobic protective material is capable of substantially preventing liquid water and/or water vapor from reaching the catalysts composition or overcoat of carbon. (Office Action, Page 5, paragraph 5.) The Office Action cites the Abstract of JP 55-023039<sup>3</sup> is cited as allegedly teaching that "the gas adsorption power of activated carbon is enhanced by treating the carbon with a water repellent resin, such as PTFE, PP or PS or a water repellent [sic] made of silicon[sic]-containing compounds, such as dimethylchlorosilane or silicone oil." (*Id.*) Appellants respectfully submit that it is incumbent upon the Examiner to supply a full translation of JP 55-023039 in the Examiner's Answer as required by MPEP § 1207.02. The extent of the teachings of the Abstract of JP 55-023039 is unclear, and it is quite possible that a translation of the full text document may include teachings away from the invention that will preclude an obviousness rejection.

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<sup>3</sup> Appellants respectfully submit that providing only the Abstract of JP 55-023039 is contrary to the requirements of MPEP § 706.02. Moreover, the Appellants previously supplied a translation of JP 52-12290 in the previously submitted Appeal Brief in this application, in response to which the finality of the last office action was withdrawn and the last Office Action was issued, which now relies on the Abstract of JP 55-023039.

The Office Action concludes that it would have been obvious for one of ordinary skill in the art to provide at least one layer of a hydrophobic protective material to overcoat the porous carbon overcoat in the modified apparatus of WO 98/02235, on the basis of suitability for the intended use, because the hydrophobic protective material prevents the condensation of moisture in the carbon pores so that the activated carbon exhibits a stable adsorption power.

Regarding claims 49 and 50, claim 49 recites a hydrophobic protective overcoat over the overcoat of carbon, the hydrophobic overcoat being capable of preventing liquid water and/or water vapor from reaching the catalyst composition or overcoat of carbon, and claim 50 recites a layer of a hydrophobic protective material coated over the catalyst composition or over the overcoat of carbon. There is no teaching or suggestion in any of the references relied upon by the Office Action to apply at least one overcoat of a hydrophobic protective material over the overcoat of carbon. The Office Action admits that WO 98/02235 and JP 52-12290 fail to teach the limitations of claims 49 and 50. As such, *prima facie* obviousness has not been established.

As noted above, replacing the coating in WO 98/02235, which is intended to resist chemicals, salts, dirt and other contaminants which may adversely affect the catalyst composition, with an adsorptive coating such as carbon would destroy the intended function of the coating in WO 98/02235. The Office Action now suggests that a person of skill in the art would further modify the modified structure in WO 98/02235 to provide a hydrophobic coating over the adsorptive coating. Appellants maintain that this further modification would still destroy the intended function of the original coating in

WO 98/02235, which is to repel chemicals, salts, dirt and other contaminants by providing an adsorptive coating of carbon having a hydrophobic coating. Hence, there would be no motivation to combine the references. See MPEP § 2143.01 Subheading V. For this additional reason, the Office Action fails to establish *prima facie* obviousness.

Moreover, reliance on the Abstract JP 55-023039 is misplaced. First, there is no teaching or suggestion in the Abstract of JP 55-023039 that the adsorptive power of the activated carbon is "enhanced" as stated by the Office Action. Second, the rejection fails to address why one of ordinary skill in the art would remove the repellant layer as taught in WO 98/02235, which is designed to repel chemicals, salts, dirt and other contaminants, replace it with an adsorptive layer and then place a hydrophobic layer over the layer of carbon. There is no teaching that a hydrophobic layer would be useful in repelling chemicals, salts, dirt and other contaminants, as required in WO 98/02235, and thus, there would be no reasonable expectation of success in making the proposed combination suggested in the Office Action. Because a reasonable expectation of success is lacking, the Office Action further fails to establish a *prima facie* case of obviousness.

Additionally, it is apparent from the Abstract of JP 55-023039 that the activated carbon is utilized for controlling harmful gas and odorous gas, and that such applications probably do not encounter high temperatures. At lower temperatures, a water repellant coating may be desired. However, the Office Action suggests combining the teachings of the Abstract of JP 55-023039 with the catalyst coatings of WO 98/02235 that encounter high temperatures. Because the catalyst in WO 98/02235 is expected to be exposed to high temperatures, the need for a water repellent coating to "prevent the condensation of moisture in the carbon pores so that the activated carbon exhibits stable absorption

power" is probably unnecessary due to the fact that moisture would probably not be problematic at high temperatures. In addition, as discussed above, WO 98/02235 seeks to provide a coating that repels chemicals, salts, dirt and other contaminants, and the skilled artisan would not want to provide an adsorptive coating as suggested combined teachings of the references cited in the Office Action. For the foregoing reasons, claims 49 and 50 are *not* obvious in view of the cited art.

**Claim 55**

For at least the reasons provided above with respect to claims 49 and 50, claim 55 is not obvious.

### **Conclusion**

In view of the foregoing, claims 48-53 and 55-58 are definite under 35 U.S.C. § 112. Furthermore, claims 48-53 and 56-58 of the application are not obvious under 35 U.S.C. § 103(a).

The undersigned was authorized by Richard A. Negin, Reg. No. 28,649, an attorney of record in the subject application, to prepare and file this Appeal Brief on behalf of the Assignee. Correspondence should be directed to Chief Patent Counsel, BASF Catalysts LLC, 101 Wood Avenue, P.O. Box 770, Iselin, NJ, 08830-0770.

Dated: February 22, 2007

Respectfully submitted,

By:       /Scott S. Servilla/  
Scott Servilla  
Registration No. 40,806  
Telephone: (732) 815-0404  
Facsimile: (732) 815-1330

BASF Catalysts LLC  
101 Wood Avenue  
P.O. Box 770  
Iselin, NJ, 08830-0770

### **Claims Appendix**

48. A device for treating the atmosphere to catalytically convert atmospheric pollutants to less harmful materials comprising an outer surface of a motor vehicle component which is exposed to a flow of ambient air, said outer surface being coated with a catalyst composition, said catalyst composition being protected with a porous overcoat of carbon.

49. The device of claim 48 further comprising at least one hydrophobic protective material overcoating the overcoat of carbon, said protective material being capable of substantially preventing liquid water and/or water vapor from reaching the catalyst composition or overcoat of carbon.

50. The device of claim 48 comprising at least one layer of the hydrophobic protective material coated over the catalyst composition or overcoat of carbon.

51. The device of claim 48 wherein the catalyst composition is selected from the group comprising base metals, precious metals as well as salts and oxides thereof and combinations thereof.

52. The device of claim 48 wherein the catalyst composition comprises manganese dioxide.

53. The device of claim 48 wherein the overcoat of carbon is selected from the group comprising activated granular carbon, carbon black, permanganate on carbon and mixtures thereof.

55. The device of claim 49 wherein the hydrophobic protective material is selected from the group comprising fluoropolymers and silicone polymers.

56. The device of claim 48 wherein said outer surface of a motor vehicle component is selected from the outer surface of radiators, air-conditioner condensers, charge air coolers, transmission coolers and inserted devices.

57. The device of claim 48 wherein said catalyst composition is selected from manganese dioxide, platinum, palladium and mixtures thereof.

58. The device of claim 48 wherein the catalyst composition is supported on a material selected from ceria, alumina, titania, silica, zirconia, and mixtures thereof.

## **Evidence Appendix**

Translation of JP 52-122290.

Japan Patent Office  
Kokai Patent Publication No. Sho 52 (1977) – 122290, October 14, 1977

Catalyst for use in the treatment of exhaust gas

Patent Application No. Sho 51 (1976) – 39069  
Date of Application: April 7, 1976

Inventor: S. Adachi  
c/o Tokyo Denki Kagaku Kogyo (Tokyo Electrochemical Industries) K. K.  
14- 6 2- Chome, Uchi Kanda, Chiyoda Ku, Tokyo  
Inventor: A. Koyama  
c/o Tokyo Denki Kagaku Kogyo (Tokyo Electrochemical Industries) K. K.  
14- 6 2- Chome, Uchi Kanda, Chiyoda Ku, Tokyo  
Applicant: Tokyo Denki Kagaku Kogyo (Tokyo Electrochemical Industries) K. K.  
14- 6 2- Chome, Uchi Kanda, Chiyoda Ku, Tokyo

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### Specification

#### (1). Title of Invention

Catalyst for use in the treatment of exhaust gas

#### (2). Claims of the Patent

[1]. Catalyst for use in the treatment of exhaust gas, the catalyst being used at the treatment of exhaust gas and characterized by that at least a part of the surface of the catalyst is covered with the layer of a porous material that is different from the catalyst component.

[2] The catalyst for use in the treatment of exhaust gas described in Claim 1 in which the material that covers at least a part of the surface of the catalyst is at least one that is selected from aluminum oxide, active carbon, alkali metal oxide, alkali earth metal oxide, the metal oxide or sulfate of the transition metal such as iron, manganese, copper, nickel, cobalt, zinc, cadmium, etc.

#### (3). Detailed Description of the Invention

This invention is related to the catalyst for use in the treatment of exhaust gas that contains nitrogen oxide. More specifically, this invention is related to the catalyst which is suitable in the treatment of exhaust gas for the effective reduction and removal of NO<sub>x</sub> in the gas that simultaneously contains nitrogen oxide (hereinafter, this is called NO<sub>x</sub> and sulfur oxide).

The pollution of atmospheric air by the NO<sub>x</sub> which is contained in the exhaust gas from automobiles and in the waste gas from the various combustion furnaces, heavy oil burners, incinerators, chemical plants, etc. are being discussed as a social problem and the appearance of an effective means of making it harmless is being desired.

Up to the present, as for the means of removing the NO<sub>x</sub> contained in the waste gas, the method of passing it to a washing solution to have it absorbed, the method of having it adsorbed to an adsorbing agent, the method of having it contact with a catalyst to convert it to a harmless material, etc. were known.

However, the washing method uses a large amount of liquid and so the handling is inconvenient and also it requires facilities of a large scale and the adsorbing method is unsuitable for the treatment of a large amount of the waste gas. For this reason, the method of treatment by using a catalyst has been drawing attention and, as the catalyst for use in this treatment of waste gas, the metals of platinum family, the oxides of metals such as iron, manganese, copper, molybdenum, vanadium, etc., or the catalyst prepared by having these carried in a carrier such as alumina, silica, etc. were proposed. However, the catalysts said above which have been known from the past are weak against the poisoning primarily by the sulfur oxide contained in the waste gas and so they have had problems in terms of the life.

The present invention eliminates the shortcomings described above and provides an inexpensive catalyst in which the catalyst component is not poisoned by the compounds of sulfur, lead, calcium, nickel and the steam, vanadium compound, nickel compound, etc in the waste gas containing nitrogen oxide and so the catalyst activity is maintained stably for a long time.

Thus, this invention is a catalyst that is used for converting harmless the nitrogen oxide in the waste gas and, by making a double structure in which a part of the catalyst surface is covered with a porous material that is different from the catalyst being used and by this, the compounds of sulfur, phosphorus, lead, etc are adsorbed selectively to the porous material to prevent the adsorption or sticking to the catalyst component and prevent the degradation of the catalyst activity. Further, the severe degradation of the strength of catalyst is prevented.

As for the porous material which is used in this invention, the examples are: aluminum oxide, active carbon or the alkali metal oxide such as Na<sub>2</sub>O, K<sub>2</sub>O, etc.; the alkali earth oxide such as CaO, SrO; the oxides of transition metal such as Fe, Mn, Cu, Ni, Co, Zn, Cd, etc., their mixture and compound; also their compounds such as sulfate.

Also, the porous material that is different from the catalyst is for protecting the catalyst by covering the catalyst surface with a material that does not necessarily have the catalyst activity based on the key point of this invention and it means to have a proper porosity so that it does not give a large hindrance to the movement of the reactant and product to and from the catalyst surface. As to the thickness of its covering, 5 μm ~ 1000 μm is suffi-

cient and, as for the covering ratio, 10 % ~ 100 % relative to the catalyst surface area is sufficient. Further, as for the catalyst component which is used in this invention, there is no particular restriction as long as it is an effective one as the catalyst for use in the treatment of waste gas, including those that are known as the catalyst for use in the treatment of waste gas such as the platinum- carrying catalyst, the sintered oxide of transition metal such as vanadium, iron, copper, nickel, cobalt, manganese, chromium, molybdenum, tungsten, etc and those in which these are carried on the carriers and those in which their sulfate is carried on carrier. This is natural in view of the key point of this invention.

As for the waste gas that is the object of treatment of this invention, there are the boiler waster gas, and the waste gas from various types of combustion furnaces, heavy oil burn-ers, incinerators, chemical plants, automobiles, etc. containing NO<sub>x</sub>,

As for the reducing gas that is added when necessary in the catalyzed reduction of NO<sub>x</sub>, there are the carbon monoxide, hydrogen sulfide, hydrogen, hydrocarbon or their mixture, or the water gas, etc.

In the following examples of application, ammonia is used as the reducing gas but, as is clear from the key point of this invention, it is not limited to this and, also, it goes without saying that the catalyst that is used is not limited to these examples of application.

By using the catalyst of this invention, for example, when ammonia is added as the reducing gas into the waste gas containing NO<sub>x</sub> and, in the temperature range of 200 ~ 600 deg C, the treatment is conducted at the spatial velocity of  $1 \times 10^3 \sim 1 \times 10^5 \text{ hour}^{-1}$  (translator's note: the power indices of 10, i.e. 3 and 5 here are not clearly legible in the original copy of the patent; check with the more clearly legible copy), conversion of the waste gas to harmless gas can be conducted for a long time with a high efficiency of cleaning. The amount of addition of the ammonia needs to be at least at 2/ 3 mol ratio with respect to the nitrogen oxide but, when a complete cleaning is not necessarily re-quired, the amount of ammonia can be reduced.

In the following, detailed explanation is given by using the examples of application.

#### Example of Application 1

In the carrier made of porous alumina of spherical diameter of 6 mm, by using the ordi-nary method of impregnation, iron oxide and vanadium oxide were carried by a ratio of Fe/ V = 7/ 3 (atomic ratio). It was confirmed that the component of the carried substance was contained by 5.0 wt % with respect to the alumina. (This catalyst is designated as A).

On the surface of the catalyst A, by using the ordinary pan type granulating machine, Fe<sub>2</sub>O<sub>3</sub> was applied and baking was done at 500 deg C to for the covering (This catalyst is designated as B.). Under a microscope observation, it was confirmed to have a double structure in which the Fe<sub>2</sub>O<sub>3</sub> layer of about 50 μm was covering the surface.

The catalysts A, B that were obtained as described above were filled in a catalyst tank and this was maintained at 300 deg C and, to this, the waste gas from a boiler which was burning the heavy oil B (NO approximately 100 ppm, SO<sub>2</sub> 150 ppm, CO<sub>2</sub> 10 %, steam 10 %, O<sub>2</sub> 4 %, remainder N<sub>2</sub>) was passed at a spatial velocity of 5000 hr<sup>-1</sup> with respect to the amount of the catalyst for 1000 hours. (These catalysts are designated as A', B', respectively).

With these catalysts A, A', B, B' and the catalyst B'' obtained by removing the Fe<sub>2</sub>O<sub>3</sub> layer of the surface of the catalysts B', 40 cc of each catalyst was filled in a reactor tube made of quartz glass and the mixed gas consisting of NO 150 ppm, SO<sub>2</sub> 150 ppm, NH<sub>3</sub> 150 ppm, O<sub>2</sub> 1.5 %, steam 10 %, and remainder of nitrogen was passed at a spatial velocity of  $5 \times 10^3$  hr<sup>-1</sup> and the NO cleaning ratio, [(NO (inlet) – NO (outlet))/ NO (inlet)] x 100, was measured. Also, the elements at the surface of these catalysts were analyzed by the fluorescent X ray analysis. From the sample after a 1000 hours test, sulfur and a small amount of phosphorus (translator's note: the phosphorus here was not clearly legible in the patent copy and it may be other substance) were detected in addition to the catalyst component. Among these, the result of measurements for the sulfur and the cleaning ratio are shown in Table 1.

Table 1

|     | Cleaning ratio at 300 deg C | S (K or primary)* |
|-----|-----------------------------|-------------------|
| A   | 80 %                        | -                 |
| A'  | 75 %                        | 600 cps           |
| B   | 78 %                        | -                 |
| B'  | 77 %                        | 650 cps           |
| B'' | 79 %                        | 30 cps            |

\* Translator's note: The word "primary" could be read as "first order" or "first time" but exactly what it means is not clear from the context. What the "cps" stands for is not clear also.

## Example of Application 2

By the same method as in Example of Application 1, to the surface of catalyst A of Example of Application 1, Mn<sub>2</sub>O<sub>3</sub> layer of about 70 μm was covered to obtain the catalyst C. This catalyst C was tested under the same condition as in Example of Application 1 and the result is shown in Table 2. Also, as in Example of Application 1, the catalyst C' is the catalyst C that was exposed to the boiler waste gas for 1000 hours and the catalyst C'' is the catalyst C' from whose surface the Mn<sub>2</sub>O<sub>3</sub> was removed.

Table 2

|     | Cleaning ratio at 300 deg C | S (K or primary)* |
|-----|-----------------------------|-------------------|
| A   | 80 %                        | -                 |
| A'  | 75 %                        | 600 cps           |
| C   | 77 %                        | -                 |
| C'  | 76 %                        | 680 cps           |
| C'' | 79 %                        | 10 cps            |

\* Translator's note: The word "primary" could be read as "first order" or "first time" but exactly what it means is not clear from the context. What the "cps" stands for is not clear also.

As is clear from these results, with A, A', the active components received the poisoning of the sulfur oxide, etc. in the waste gas component and so the activity degraded while, with the catalyst B, B', B'', C, C', C'', the component of sulfur oxide, etc. (believed to be mainly SO<sub>2</sub>) was substantially adsorbed to the Fe<sub>2</sub>O<sub>3</sub>, Mn<sub>2</sub>O<sub>3</sub> that covered the surface and so it did not reach the catalyst component and thus it is seen that there was less degradation of the catalyst activity. Also, when other catalyst component was used as in Example of Application 1 and the test was conducted on the catalyst of this invention that had the double structure covered with various porous materials which were different from the catalyst component, good results were obtained similarly.

Continuation of p. 1

Inventor: M. Takayama  
c/o Tokyo Denki Kagaku Kogyo (Tokyo Electrochemical Industries) K. K.  
14- 6 2- Chome, Uchi Kanda, Chiyoda Ku, Tokyo

## **Related Proceedings Appendix**

None.